

The new isotopes of element 107: Bh-266 and Bh-267 and the chemistry of bohrium

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The early history of bohrium

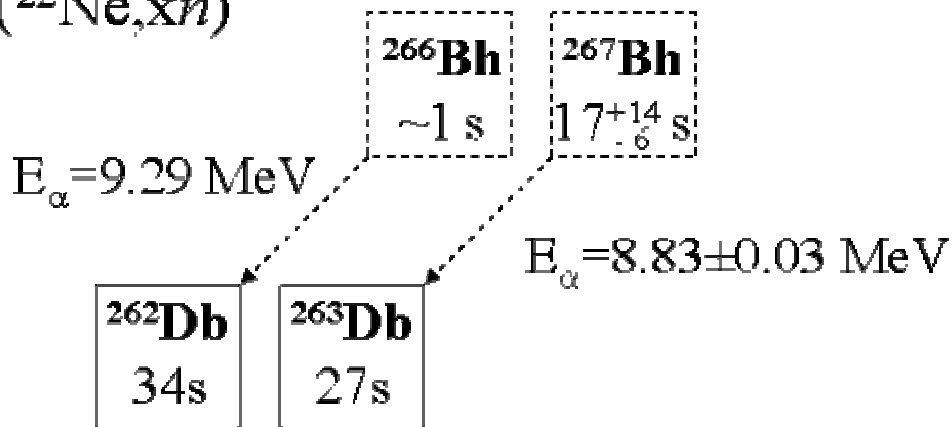
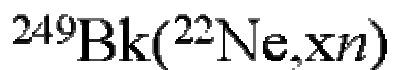
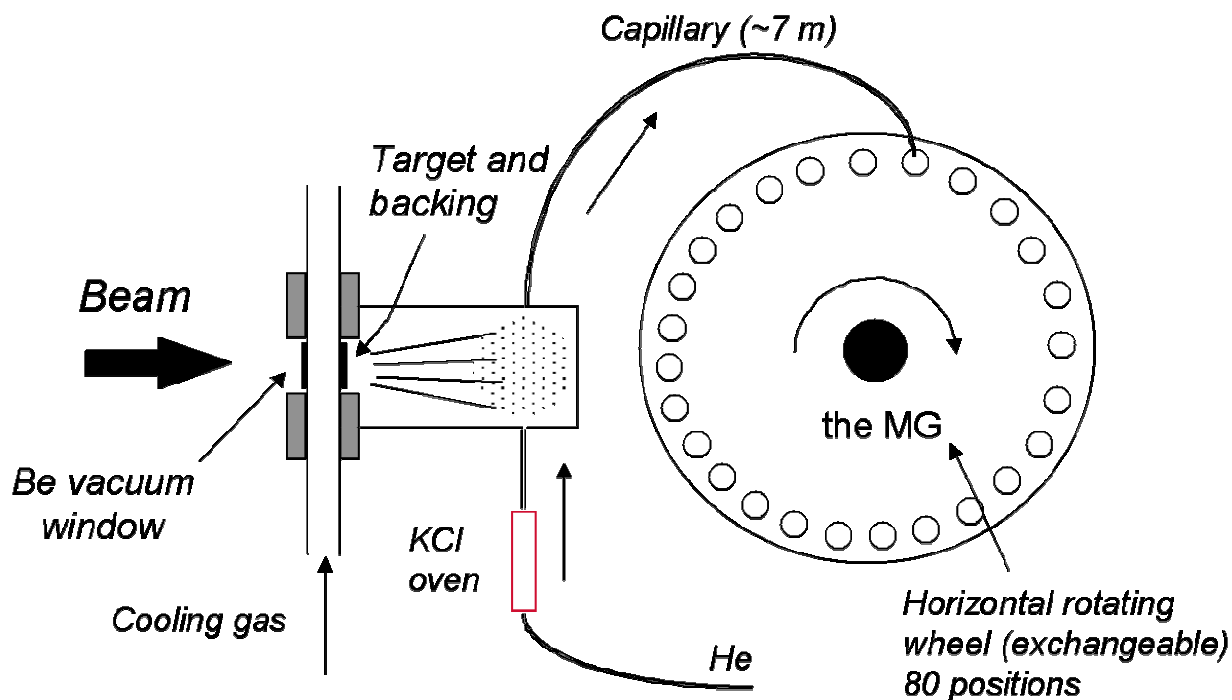
Bohrium was first identified in 1981 by Münzenberg *et al.* as the isotope $^{262}\text{Bh}^m$ produced in the $^{209}\text{Bi}(^{54}\text{Cr},n)$ reaction [1] using the velocity filter “Separator for Heavy Ion Production” (SHIP) at the Gesellschaft für Schwerionenforschung (GSI), Darmstadt, Germany. Earlier performed experiments initially claimed synthesis of element 107 via the $^{54}\text{Cr}(^{209}\text{Bi},2n)$ reaction, based on observation of the spontaneous fission activity from the decay of ^{261}Bh . However, in subsequent experiments spontaneous fission activity was not detected, and ^{261}Bh was unambiguously identified by correlation with its α -decay daughter ^{259}Ha . [2]. To date, no spontaneous fission activity has been observed that can be attributed to spontaneous fission of any bohrium isotopes and a low upper limit on the spontaneous fission branch was set. The properties of the isotopes of bohrium known previous to this work are: ^{261}Bh ($T_{1/2}=11.8$ ms; $E_{\alpha}=10.40, 10.10, 10.03$ MeV [2]), ^{262}Bh ($T_{1/2}=102$ ms; $E_{\alpha}=10.06, 9.91, 9.74$ MeV [2]), $^{262}\text{Bh}^m$ ($T_{1/2}=8$ ms; $E_{\alpha}=10.37, 10.24$ [2]), and ^{264}Bh ($T_{1/2}=440$ ms; $E_{\alpha}=9.62, 9.48$ MeV [3]).

The isotopes 266 and 267

Experiments were undertaken to try to produce and identify the new neutron-rich isotopes of bohrium, ^{266}Bh and ^{267}Bh . These isotopes were predicted to have significantly longer half-lives than previously known bohrium isotopes, possibly long enough to enable the first studies of bohrium chemical properties in subsequent experiments. Previous attempts to identify these isotopes by chromatographic separation have failed [4, 5]. Based on predicted Q -values for electron capture and α -decay, ^{266}Bh and ^{267}Bh should decay predominantly by α -emission and possibly by spontaneous fission (SF). The α -particle energies and half-lives for these isotopes are expected to be in the range of 8.7–9.3 MeV [6] and 1-20 seconds [7]. The previously reported decay characteristics of their Ha and Lr daughter nuclei, are: ^{262}Ha ($T_{1/2}=34$ s; $E_{\alpha}=8.45, 8.53, 8.67$ MeV [8]), ^{263}Ha ($T_{1/2}=27$ s; $E_{\alpha}=8.35$ MeV [9]), ^{258}Lr ($T_{1/2}=3.9$ s [10]; $E_{\alpha}=8.60, 8.62, 8.57, 8.65$ MeV [8]), and ^{259}Lr ($T_{1/2}=6.34$ s; $E_{\alpha}=8.45$ MeV [10]). Kratz *et al.* [9] report an α -particle energy of 8.35 MeV for ^{263}Ha , but other measurements [11] indicate that ^{263}Ha might also decay by emission of 8.41-MeV α -particles.

Using these predictions as a guide, ^{267}Bh and ^{266}Bh were produced in bombardments of a ^{249}Bk target with 117-MeV and 123-MeV ^{22}Ne ions. Identification was made by observation of correlated α -particle

decays between the bohrium isotopes and their daughters by using our rotating wheel (MG) system. A parent-daughter stepping mode was used to provide detection of α - α correlations with a greatly reduced background [12]. After the end of bombardment and careful analysis of the data, it was found that four atoms of ^{267}Bh were produced during the experiment. ^{267}Bh was produced with a cross-section of ~ 70 pb and was found to decay with a $17 (+14/-6)$ s half-life by emission of α particles with an average energy of 8.83 ± 0.03 MeV. One atom of ^{266}Bh was observed, decaying within one second by emission of a 9.29-MeV α particle [13].



The chemistry of bohrium

The arrangement of the chemical elements in the periodic table indicates similarities of chemical properties, which reflect the elements' electronic structure. For the heaviest elements, however,

deviations in the periodicity of chemical properties are expected due to relativistic effects. Bohrium is expected to be a group seven element and should form oxychloride species in the gas phase in an analogous fashion to rhenium and technetium. During test experiments with rhenium and technetium [14, 15] only one chemical species, assumed to be $[\text{Re/Tc}]\text{O}_3\text{Cl}$, was seen with the thermochromatography apparatus.

Pershina *et al.* predict that the electronic structure of BhO_3Cl should be very similar to TcO_3Cl and ReO_3Cl . The calculated energy gap (ΔE) and ionization potential (IP) increase from Tc to Bh for the MO_3Cl species. The 7+ oxidation state should also increase in stability down the group following the increasing ΔE . The Bh species is also more covalent than the other two species which they attribute to relativistic effects seen in the component atomic orbital [16, 17]. These effects also influence dipole moments and dipole polarizabilities, which should dominate intermolecular or molecule-surface interactions. These interactions are the basis for volatility that they expect should decrease down the group, i.e. that BhO_3Cl should be less volatile than ReO_3Cl which should be less volatile than TcO_3Cl .

Using the same reaction as for the previous experiment, a volatility experiment to study bohrium chemistry was performed at the Paul Scherrer Institute (PSI), Switzerland. The behavior of Bh was investigated at 180, 150, and 75°C. Two α -SF chains, two α - α chains, and one α - α - α decay chain, all attributed to the decay of ^{267}Bh , were observed at 180°C. At 150°C two α -SF chains were observed, and at 75°C no events were observed. Random correlations should not significantly contribute to the number of α -SF or α - α - α chains seen, but approximately one of the α - α correlations seen are expected to be due to random correlations. The unambiguous identification of Bh after chemical separation allows us to conclude that like its lighter homologues, Bh forms a volatile oxychloride compound, presumably BhO_3Cl , and behaves like a typical group seven element [18]. We found that this compound is less volatile than the oxychlorides of the lighter elements of group VII, thus confirming predictions based on its periodic position.

Assuming the Bh compound to be BhO_3Cl and applying a microscopic model of the adsorption-desorption process [19] and using the Monte Carlo code developed by Türlér to fit the measured volatility curves, the evaluated standard adsorption enthalpy (ΔH_{ads}) of BhO_3Cl on the quartz surface was determined to be $-75(+9/-6)$ kJ/mol. The adsorption enthalpies for TcO_3Cl and ReO_3Cl are -51 ± 3 and -61 ± 3 kJ/mol respectively [15]. After the quartz column, BhO_3Cl was transported to ROMA on CsCl aerosols. Unlike ReO_3Cl , TcO_3Cl is so volatile that it can not be transported with CsCl, which again indicates Bh is more like Re than Tc.

Conclusion

Bohrium has been placed decisively on the periodic table and periodic extrapolations confirmed. This first chemistry experiment with bohrium would have been impossible without an adequate candidate isotope of bohrium. These experiments were also reliant on the theoretical predictions of the chemical properties of bohrium and the theoretical predictions of the production and decay properties of the anticipated heavier isotopes of bohrium. These experimental data that we have generated, can now be re-applied to the theory to further fine-tune and increase our understanding of the fundamental processes at work [20,21]. In addition, the target material was provided by Office of Science, Office of Basic Energy Research, Division of Chemical Sciences, of the U.S. Department of Energy, through the transplutonium element production program at the Oak Ridge National Laboratory. The culmination of this collaboration, by the successful determination of the chemistry of bohrium, was only possible by the joint efforts of all the members from many different organizations.

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